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Initial flux of sediment-associated radiocesium to the ocean from the largest river impacted by Fukushima Daiichi Nuclear Power Plant

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This study aimed to quantify the flux of radiocesium in the Abukuma Basin (5,172 km²), the largest river system affected by fallout from the Fukushima Daiichi Nuclear Power Plant (FDNPP) event. In the period from 10 August 2011 to 11 May 2012 an estimated 84 to 92% of the total radiocesium transported in the basin's fluvial system was carried in particulate form. During this monitoring period Typhoon Roke (September 2011) was observed to induce a significant and temporally punctuated redistribution of radiocesium. The storm-mobilised radiocesium was an estimated 6.18 Terabecquerels corresponding to 61.4% of the total load delivered to the coastal zone during the observation period. The total flux of radiocesium into the Pacific Ocean estimated at the outlet station (basin area 5,172 km²) was 5.34 TBq for ¹³⁷Cs, and 4.74 TBq for ¹³⁴Cs, corresponding to 1.13% of the total estimated radiocesium fallout over the basin catchment (890 TBq). This was equivalent to the estimated amount of direct leakage from FDNPP to the ocean during June 2011 to September 2012 of 17 TBq and the Level 3 Scale Leakage on 21 August 2013 (24 TBq).

Transfer of sediment from land to the ocean by rivers is a key pathway for material transfer on Earth¹. River basins contribute an estimated 13 Gt of sediment per year to the world's coastal zone² and the affinity of many contaminants for fine sediment³ means that river networks are an important conveyance route for transfer of terrestrial contaminants to coastal and marine ecosystems. In this context, recent contamination of the land surface by radioactive fallout in the vicinity around the Fukushima Daiichi Nuclear Power Plant is of much concern. While direct inputs of radionuclides from FDNPP to coastal waters have been estimated and modeled⁴, less is known about the flux of radionuclides to the coastal zone derived from radioactive runoff into the river basin networks. Indeed, soil inventories of radionuclides in the river basin are reported to be significantly elevated^{5,6} with implications for terrestrial ecosystem function. The main vector for radiocesium transport in runoff has previously been attributed to suspended sediments^{7,8}. Importantly, extreme weather events have led to punctuated remobilization of significant quantities of radionuclides into river systems. In storm events the volume of water entering the fluvial system over a short period is significantly increased, accordingly increasing the mass and size of the suspended particulate load. In the present study we report the initial flux of sediment-associated radiocesium to the coastal zone in the Abukuma River, the largest river basin impacted by FDNPP fallout.

The redistribution of radionuclides through hydrological processes (hereafter referred to hydrological redistribution) is hypothesized to pose a critical threat to aquatic and marine ecosystem health especially in downstream regions where direct contamination through fallout was absent. In this context, sediment storage zones will potentially become sink zones for radionuclides after a nuclear accident. Hence floodplains and riparian zones that did not receive direct fallout might acquire significant radiocesium inventories as a consequence of inundation and deposition of contaminated sediment from upstream source areas.

Hydrological redistribution of radionuclides within river basins was first investigated in 1960's for catchments affected by fallout from the nuclear weapons testing. However only after the Chernobyl disaster in 1986 were the effects and consequences studied in detail. Vast areas of Europe were contaminated by fallout from Chernobyl in which most affected areas was associated with rain from a single weather system⁹. Contamination of soil and the subsequent risk of remobilization and redistribution of radionuclides post-Chernobyl has now been explored in a range of landscapes. For example, IAEA¹⁰ summarized radiological conditions in the Dnieper River basin, following the Chernobyl incident, located upstream of the city of Kiev, Ukraine. In this report, transport of fallout (primarily ⁹⁰Sr and ¹³⁷Cs) was evaluated for the Dnieper Reservoir systems, in which the majority of radiocesium was determined to be associated with clay-sized particles. Here the main potential threat was considered to be a theorized catastrophic release of radionuclides into the heavily populated region downstream of the Kiev Reservoir dam. A modeling approach was adopted using "AQUASCOPE", a simplified dynamic model to predict radiocesium, radiostrontium, and radioiodine concentrations in river basin sediments and fishes. Using this approach Smith *et al.*¹¹ determined model outputs for the Kiev Reservoir and Chernobyl cooling pond using parameterization data derived from several monitoring sites situated across since the end of the atomic testing era and following the Chernobyl accident.

Similar hydrological redistribution of radiocesium was observed to have occurred in the Techa River Basin in the South Ural region of Russia, a highly contaminated river basin associated with the Kyshtym disaster that occurred at the Mayak nuclear fuel reprocessing plant in 1957. Although half a century after the Kyshtym disaster, Kryshev *et al.*¹² reported results of a numerical modeling approach for determining radiological distribution and impact in the Techa River Basin. Quantification of hydrological redistribution of radiocesium was attempted through numerical simulation of the Techa River, which is considered to be one of the most significant river basin study areas for investigating the effect of radiological pollution on an aquatic ecosystem. The South Ural region is notorious for its dry winters and relatively low-levels of annual precipitation (300–400 mm). Accordingly it does not provide a comparable analogue to the Fukushima prefecture which is located in a monsoon zone and experiences much higher levels of annual precipitation. The fallout and redistribution of radionuclides in the region surrounding FDNPP is the first example of its kind in a monsoon region. Correspondingly the monitoring in Abukuma River Basin (Figure 1), the largest river basin situated within the contaminated zone, is of significant importance.

Continuous direct release of ¹³⁷Cs into the ocean has recently been discussed by Kanda (2013)⁴ where the estimated release has been established through comparing published data of radionuclide concentrations in the artificial harbor and surrounding ocean. It was estimated that the total radionuclide released into the Pacific Ocean from 1 June to 30 September 2012 was 17.1 TBq.

Recent work by Nagao *et al.*⁷ in the Natsui and Same River basins of the Fukushima prefecture (749 and 600 km², respectively) demonstrated (i) the significant role of extreme weather events in the smaller river basin and (ii) the importance of sediment as a primary transport vector for radiocesium transfer. They also estimated the approximate flux into the ocean for these two small river basins and concluded that the total flux during the 305 day monitoring period was in the order of 0.06 TBq.

In the present study area, following the FDNPP disaster, a comprehensive research program was initiated to evaluate the transfer of radioactive fallout between the atmosphere, biosphere and hydrosphere. Within the Fukushima contaminated zone, several researchers have reported the current status of soil and biota. Kato *et al.*⁵ surveyed the vertical distribution of ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs in forested

soil profiles. Kato *et al.*⁶ also performed a detailed survey of radionuclide distribution in coniferous forest canopies.

The present study aimed to quantify the longitudinal flux of particulate-associated radiocesium (¹³⁴, ¹³⁷Cs) from the 5,172 km² catchment of the Abukuma River Basin to the coast. The following objectives were undertaken: (i) high resolution measurement of flow and suspended sediment concentrations at a series of nested monitoring stations; (ii) collection of time-integrated suspended sediment samples at each site to determine the activity concentration of radiocesium and (iii) combination of these data to calculate the radiocesium loads and net flux to the coastal zone. Coastal outflow monitoring locations provided a measure of the radiocesium flux to the Pacific. Measurements comprising high resolution flow and turbidity monitoring coupled with time-integrated bulk suspended sediment sampling span the period from June 2011 to May 2012. The detailed monitoring activity is described in detail in the following sections.

Results

Background condition. During the course of the observation period at the Iwanuma observation point, a significant peak in discharge was observed during the Typhoon Roke storm event in September 2011, and subsequent peaks were found during the snow-melting seasons from the end of April to the beginning of May 2012. Sediment concentration increases were observed in association with these periods of elevated discharge. Consequently the total sediment flux was concentrated during the aforementioned period.

Bulk suspended sediment samples were determined to be highly contaminated, ranging from 3.8 to 44 kBq kg⁻¹ for ¹³⁷Cs and from 3.3 to 34 kBq kg⁻¹ for ¹³⁴Cs. Comparing this with a relatively low concentration in water (from 0.08 to 0.5 Bq kg⁻¹ (L)), we may conclude that the radionuclides detected at the basin monitoring points were primarily associated with suspended particulate load.

According to observations, the mean diameter of particulates trapped in the sediment samples varied from 10–50 μm, and 10–40 μm in the downstream portion of the river. Figure 2 shows the particle size distribution of sediments trapped by the samplers at the 6 basin observation points on 9 August 2011.

Sediment-associated radiocesium flux. The total flux of radiocesium to the Pacific coast estimated at the Iwanuma station for the 274 day period from 10 August 2011 to 10 May 2012 was 5.34 TBq for ¹³⁷Cs, and 4.74 TBq for ¹³⁴Cs, a resulting 10.1 TBq radiocesium in total. Figures 3A and 3B illustrate the monthly transition of the total quantity of radiocesium transported via suspended particulate matter to each observation point, including the specified period encompassing Typhoon Roke. During this 8 day hydrological extreme event from 19–27 September 2011, the total flux of ¹³⁷Cs was 3.21 TBq, corresponding to 60.1 percent of total radiocesium load determined for the 10 month study period, and ¹³⁴Cs was 2.98 TBq, 62.9 percent respectively. These results indicate the importance of extreme hydrological events in controlling the fluxes of sediment and associated contaminants to the coastal zone⁸.

Fluxes of suspended particulate matter and associated ¹³⁴Cs and ¹³⁷Cs for the six Abukuma River basin monitoring stations are summarized in Table 1. The six stations are arranged in order of increasing catchment size (7.6 to 5,172 km²), which corresponds to increasing flow, suspended sediment and radiocesium transfer (Table 1). The duration of monitoring was similar for the Kuchibuto and Mizusakai sites (327 to 329 days), whereas the two sites along the main channel of the Abukuma River were monitored for less time (276–279 days). For the purpose of longitudinal comparison between sites, the maximum period of synchronous monitoring between all sites was used (i.e. 10 August 2011 to 11 May 2012).

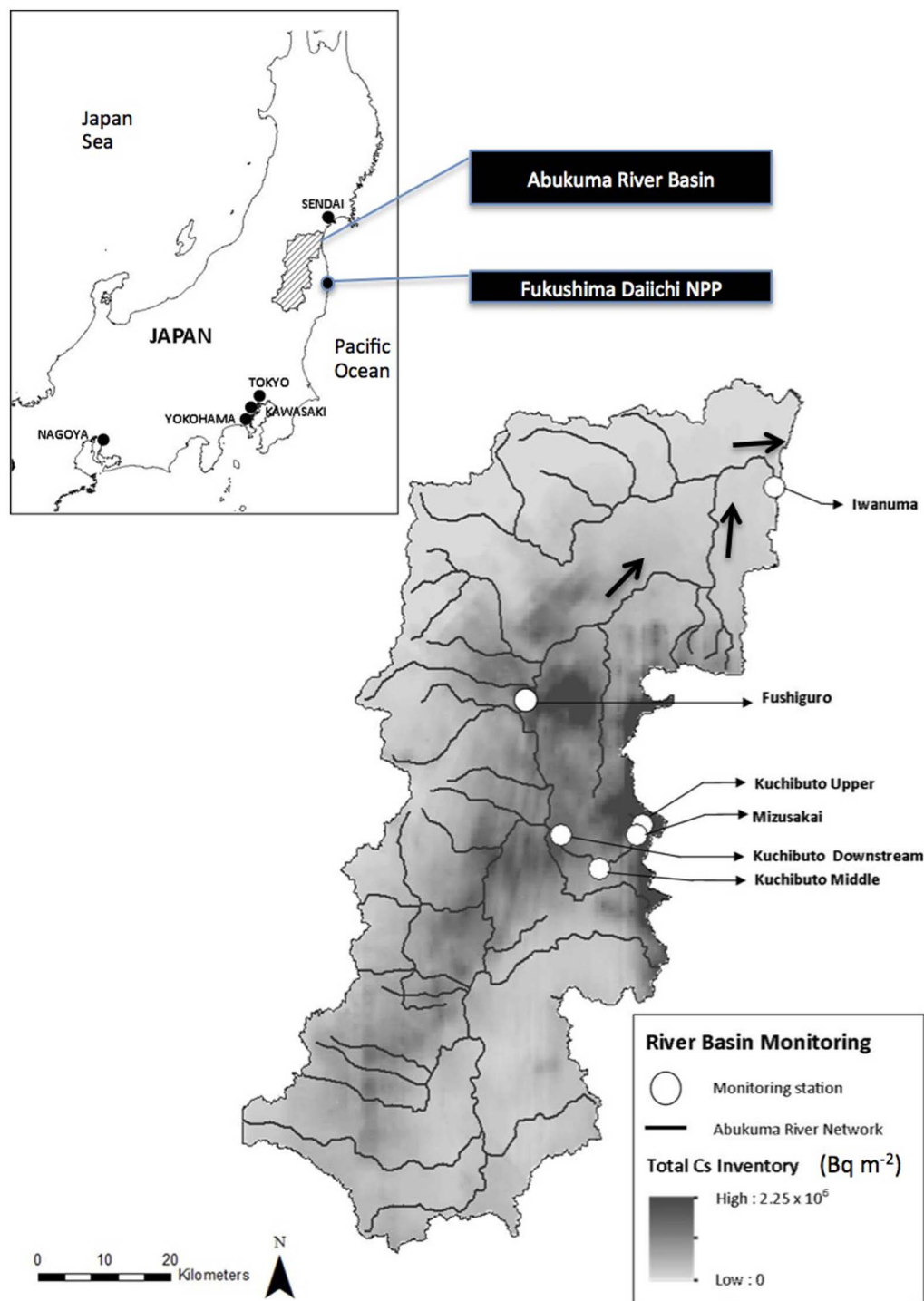


Figure 1 | Map of the Abukuma river basin showing monitoring locations and the total radiocesium inventory (maps were generated by using ArcMap 10.1 (ESRI) and GIS data obtained from Japan Map Center).

Highly elevated fluxes of ^{134}Cs and ^{137}Cs were recorded during the monitoring period (Table 1). Total radiocesium ($^{134}\text{Cs} + ^{137}\text{Cs}$) fluxes associated with suspended particulate matter ranged between 0.015 (Mizusakai) and 9.21 (Iwanuma) TBq. ^{137}Cs mostly represented a slightly larger proportion of total Cs flux (50–54%) and the load-weighted $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio ranged between 0.85–1.02, partly reflecting the more rapid decay of ^{134}Cs during the monitoring period given that the initial fallout ratio was approximately 1.0.

For the period of synchronous monitoring across all sites (10 August 2011–11 May 2012), suspended sediment fluxes ranged from 2.73×10^2 to 5.51×10^5 t. This equates to a range in suspended

sediment yields from 36 to 144 t km^{-2} . Within the Kuchibuto sub-basin, yields increased downstream and peaked at the Kuchibuto middle site (124 t km^{-2}) before decreasing at the Kuchibuto downstream site (100 t km^{-2}). Along the main channel of the Abukuma River suspended sediment yields decreased from 144 t km^{-2} to 107 t km^{-2} between Fushiguro and Iwanuma. This decrease is assumed to reflect either (i) increasing channel sediment storage in the lower reaches of the basin, especially represented by the Abukuma tidal dam located upstream of Iwanuma point, or (ii) a proportionally lower input of suspended sediment from downstream tributaries relative to the increase in basin area.

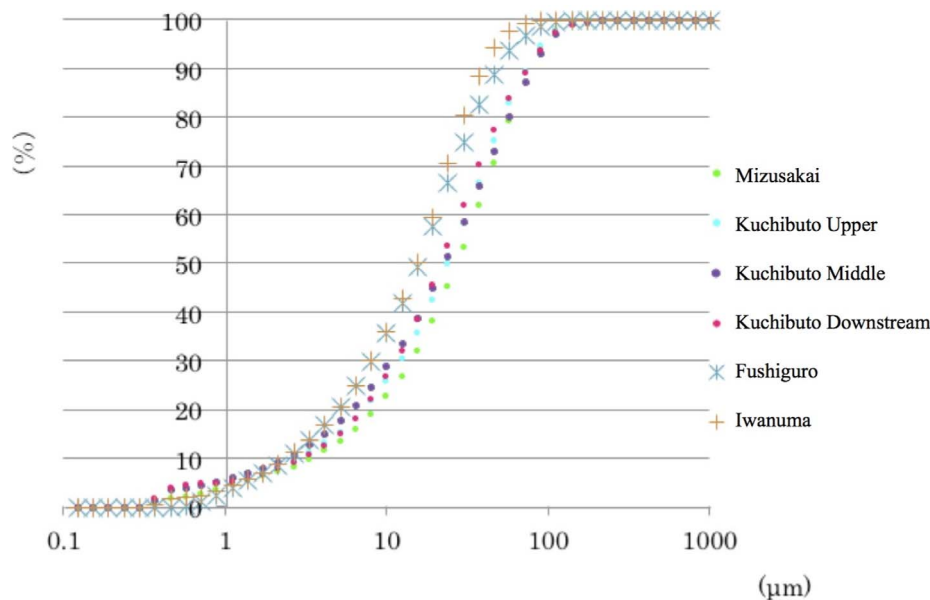


Figure 2 | Distribution of particle diameters trapped by the sediment samplers at the 6 observation points in the Abukuma River Basin on 9 August 2011.

The downstream trend in total Cs yields (kBq m^{-2}) reaches a maximum at Kuchibuto middle (2.2 kBq m^{-2}) before declining to 1.8 kBq m^{-2} at the Iwanuma station. This does not correspond to the spatially-averaged catchment inventories, which are highest for the headwaters (at Mizusakai and Kuchibuto-upper stations). Downstream changes in the particle size of suspended material may be important for interpreting these trends. In contrast, load-weighted activity concentrations (based on total Cs flux/total suspended sediment flux) correlate well with spatially-averaged catchment inventories ($R^2 = 0.90$). The highest mean activity concentrations occur in the headwaters (at Kuchibuto-upper and Mizusakai stations). The mean activity concentration decreases at Kuchibuto middle, suggesting dilution by less contaminated sediment before increasing again at Kuchibuto downstream. The Abukuma stations exhibit the lowest activity concentrations, which increase downstream from Fushiguro to Iwanuma. We neglected, however, the following information as (1) transport of radiocesium through bed-load (2) vertical distribution of suspended sediment particle concentration (3) temporal variation of radiocesium concentration in the water, and (4) behavior of radiocesium in the estuary zone.

Discussion

Only a small fraction of the total radiocesium fallout over the Abukuma River basin was transported to the coastal zone during the measurement period. This was estimated at 1.13% of total fallout over the basin based on an inventory of 890 PBq calculated from MEXT radiocesium inventory mapping¹³. The primary transport pathway in the fluvial systems has been via suspended particle load. Transport via suspended particles was estimated to account for 82 to 93% of radiocesium in the upper stream and 84 to 92% in the river mouth. During the hydrological extreme event of Typhoon Roke, an estimated 61% of the total radiocesium load recorded during the measurement period was exported from the river basin.

Our findings provide important information to assist the accurate evaluation of ecological impact and in providing countermeasures against radiocesium fluxes into the ocean from contaminated river systems. High concentrations of radiocesium were observed in sediments at the outlet of the basin, near the Abukuma river mouth, reaching 1.4 kBq/kg-dw at the river mouth in March 2013 and 0.73 kBq/kg-dw in sediments 2.4 km offshore where fine argilliferous

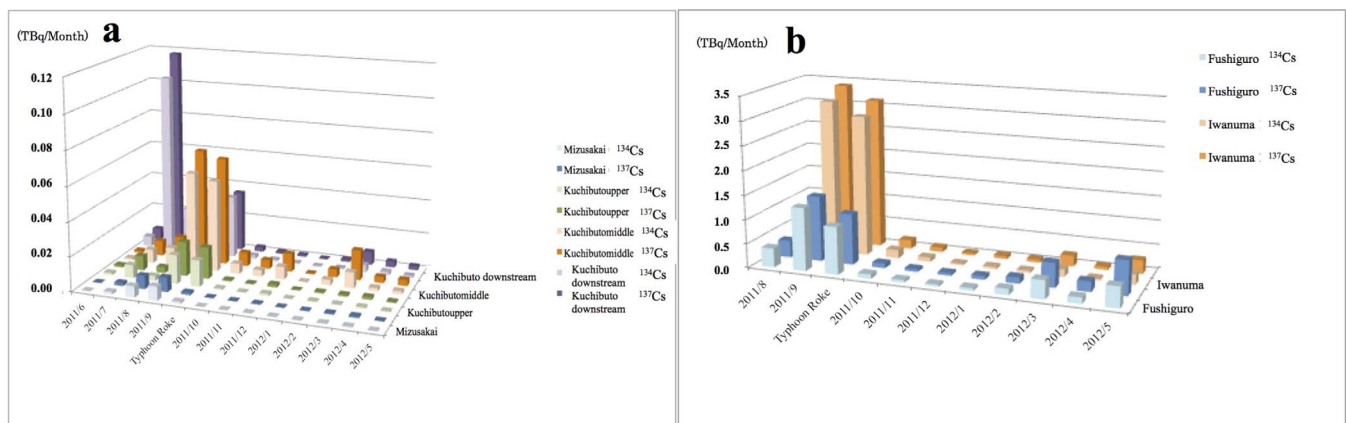


Figure 3 | Monthly variation of Radiocesium flux in (a) Kuchibuto River Basin, tributary of Abukuma River Basin, and in (b) Abukuma main stream at Fushiguro and Iwanuma Stations.

Table 1 | Summary of the fluxes of suspended particulate matter and associated ^{134}Cs and ^{137}Cs for the six Abukuma River basin monitoring stations

Abukuma River Basin monitoring: totals first year (up to May 2012)										
Monitoring station	Analysis period		Duration (d)	Total discharge (Ml)	Total suspended sediment yield (t)	Mean $\text{Cs-134} + \text{Cs-137}$ activity concentration (Bq kg^{-1})	Radiocesium flux			
	Start date	End date					Cs-137 (MBq)	Cs-134 (MBq)	Total Cs (MBq)	Total Cs (GBq)
Mizusakai	2011/6/21	2012/5/13	327	6.83E + 03	3.72E + 02	4.03E + 04	7.66E + 03	7.33E + 03	1.50E + 04	15
Kuchibuto upper	2011/6/21	2012/5/15	329	1.83E + 04	1.70E + 03	3.31E + 04	2.80E + 04	2.81E + 04	5.61E + 04	56.1
Kuchibuto middle	2011/6/21	2012/5/14	328	4.58E + 04	9.53E + 03	1.97E + 04	9.63E + 04	9.16E + 04	1.88E + 05	188
Kuchibuto downstream	2011/6/21	2012/5/14	328	7.69E + 04	1.77E + 04	2.85E + 04	2.51E + 05	2.56E + 05	5.06E + 05	506
Abukuma- Fushiguro	2011/8/9	2012/5/14	279	3.54E + 06	5.11E + 05	1.30E + 04	3.60E + 06	3.04E + 06	6.64E + 06	6,640
Abukuma- Iwanuma	2011/8/10	2012/5/11	276	5.32E + 06	5.51E + 05	1.67E + 04	4.83E + 06	4.39E + 06	9.21E + 06	9,213
Synchronous measurement period (August 2011 to May 2012)										
Monitoring station	Analysis period		Duration (d)	Total discharge (Ml)	Total suspended sediment yield (t)	Mean $\text{Cs-134} + \text{Cs-137}$ activity concentration (Bq kg^{-1})	Radiocesium flux			
	Start date	End date					Cs-137 (MBq)	Cs-134 (MBq)	Total Cs (MBq)	Total Cs (GBq)
Mizusakai	2011/8/10	2012/5/11	276	5.93E + 03	2.73E + 02	3.82E + 04	5.41E + 03	5.02E + 03	1.04E + 04	10
Kuchibuto upper	2011/8/10	2012/5/11	276	1.56E + 04	1.39E + 03	2.39E + 04	1.74E + 04	1.58E + 04	3.32E + 04	33
Kuchibuto middle	2011/8/10	2012/5/11	276	3.90E + 04	7.81E + 03	1.81E + 04	7.47E + 04	6.68E + 04	1.42E + 05	142
Kuchibuto downstream	2011/8/10	2012/5/11	276	6.52E + 04	1.35E + 04	1.93E + 04	1.37E + 05	1.23E + 05	2.61E + 05	261
Abukuma- Fushiguro	2011/8/10	2012/5/11	276	3.50E + 06	5.05E + 05	1.31E + 04	3.57E + 06	3.03E + 06	6.60E + 06	6596
Abukuma-Iwanuma	2011/8/10	2012/5/11	276	5.32E + 06	5.51E + 05	1.67E + 04	4.83E + 06	4.39E + 06	9.21E + 06	9,213

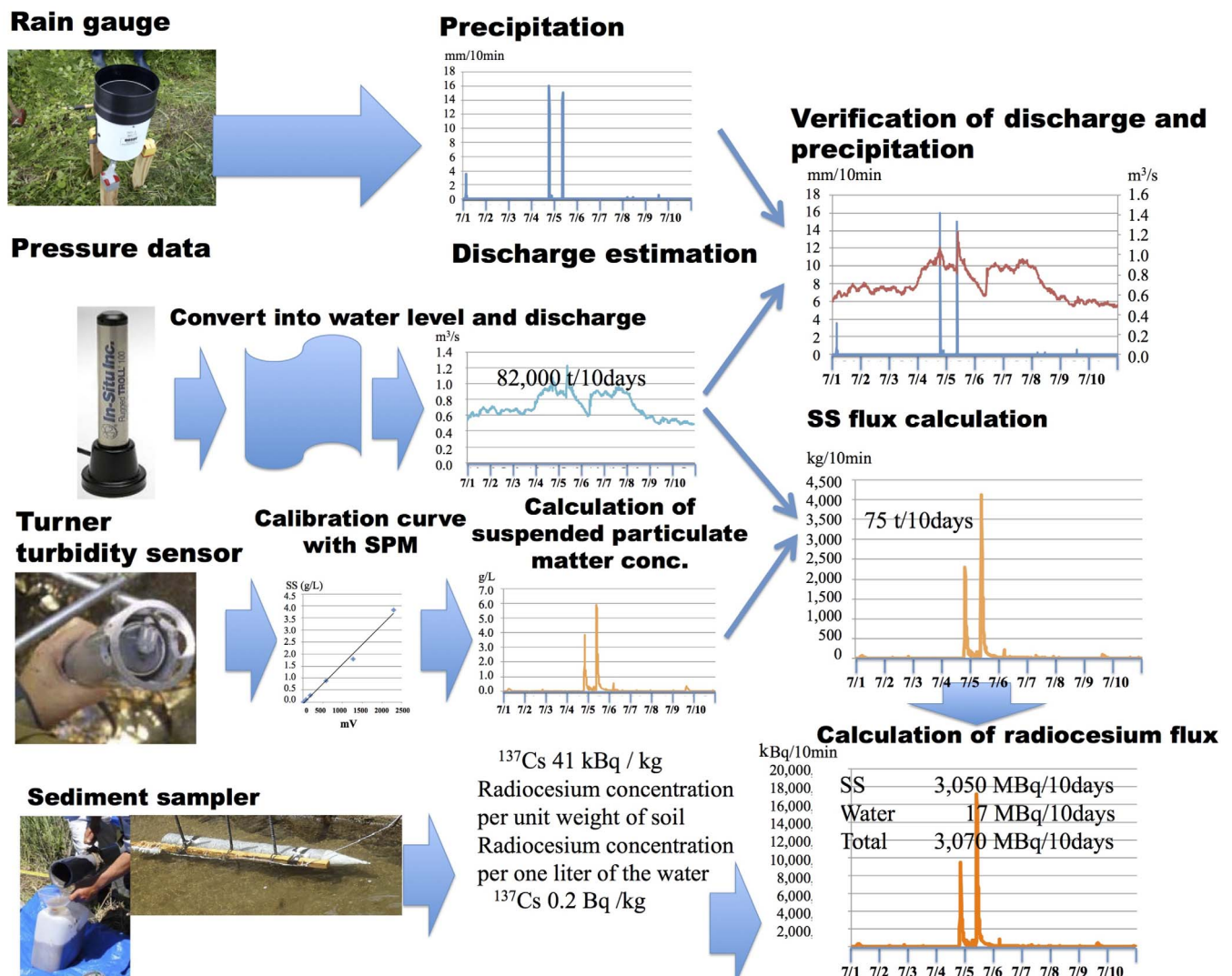


Figure 4 | Description of overall procedure in estimating radiocesium flux.

deposits are found¹⁴. Activity concentration were much higher than that in bordering coastal areas, providing important evidence of localised radiocesium contamination in basin outflow sediments from the Abukuma River. Most radiocesium attached to suspended sediment in river water can be dissolved in ambient water after reaching the estuary, due to the increase in competing ions such as sodium and potassium through mixing with seawater. On the other hand, some radiocesium in the terrestrial suspended sediment is tightly retained at the inter layer (especially illite frayed edge sites) and is rarely dissolved. Such tightly retained radiocesium may contribute to the higher activity in coastal sediment around Abukuma River mouth^{15,16}. Although radiocesium in particulate form is not considered as harmful as that occurring in dissolved form from direct leakage at FDNPP, it may be accumulated in benthic marine organisms, including several fished species typical to Sendai Bay.

We propose that greater attention should be paid to the effect of extreme storm events in rapidly and significantly increasing flux of radiocesium for short periods. Our study also determined that the total quantity of radiocesium redistributed in the river basin (10.1 TBq) is of the same order of magnitude estimated for direct leakage from the FDNPP site between 1 June 2011 to 30 September 2012⁴(estimated as 17 TBq) and for the Level 3 Scale Leakage incident occurring in August, 2013¹⁷(estimated as 24 TBq).

Sediment control is accordingly important to prevent significant downstream accumulation and transport of radionuclides into the Pacific Ocean.

Methods

The Abukuma River is the largest basin affected by radioactive fallout from the FDNPP accident. A series of nested monitoring stations were established within this basin. Two stations were installed on the main river channel at Fushiguro and Iwanuma (Figure 1). The Iwanuma site provided a measure of suspended sediment and radiocesium flux to the Pacific coast. Upstream of the Fushiguro station, the more highly-contaminated Kuchibuto sub-basin was selected for detailed monitoring. Within this sub-basin, the average spatial radiation rate varies from 3 to 10 $\mu\text{Sv h}^{-1}$ and the corresponding mean concentration of radionuclides in the soil varies around 20 to 100 kBq kg^{-1} .

At each monitoring station a Troll pressure sensor was installed to measure water level alongside a turbidity sensor. In addition, a time-integrated suspended sediment sampler based on the design by Phillips et al.¹⁸ was installed to collect bulk sediment samples for analysis. Using channel cross-sectional surveys and measured flow velocities, water levels were converted to discharge at each station. Turbidity sensor measurements (mV) were correlated with suspended sediment concentrations (SSC) to enable conversion from turbidity to SSC using a calibration curve developed through precise calibration with Kaoline. Multiplication of SSC and flow enabled the calculation of suspended sediment flux. Flow and SSC time-series data were analyzed and corrected using the US Army Corps of Engineers HEC-DSSVue software. The whole process is shown in Figure 4.

Time-integrated sampling of suspended sediment produced sufficient quantities of sediment for analyses of radiocesium (¹³⁴Cs and ¹³⁷Cs) activity concentration using high purity germanium (HPGe) gamma spectrometers, together with analysis of

particle distribution. Sediment sampling intervals varied across monitoring stations but were typically 2–4 weeks. Suspended sediment accumulated in the sediment sampler during two to four weeks was collected as turbid water as shown in Figure 4, and dried for 24 hours at temperature at 105°C. The dried sample was disaggregated using mortar and then analyzed. The measured activity concentrations (in Bq kg⁻¹) of ¹³⁴Cs and ¹³⁷Cs for each sediment sample were multiplied by suspended sediment flux to determine the flux of ¹³⁴Cs and ¹³⁷Cs for each sediment sampling interval. Radiocesium concentration in river water is calculated based on the field observation in August 2011 (averaged three consecutive observation results both in the Abukuma mainstream and Kuchibuto Basin, and by adjustment considering the decay of both radionuclides). Water samples were filtered by membrane filter (pore size of 0.45 µm). Some of filtrate (1–10 L) was mixed with ammonium molybdophosphate (AMP) to concentrate dissolved radiocesium. Radiocesium activities in the filtrate and AMP sample were measured using HPGe gamma spectrometers in Meteorological Research Institute of Japan Meteorological Agency (MRI-JMA). Radiocesium activity concentrations were decay corrected to the mid-point of each sampling interval. The analysis is based on the assumption that the time-integrated sediment samples provide an average activity concentration that may be used in conjunction with the time-varying suspended sediment flux to compute radiocesium flux for a given sediment sampling interval.

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Author contributions

Y.Y., H.S. & W.B. wrote main components of the article based on their observations and analytical results. Y.O. made field monitoring design & installation, and gave advices. T.W. and Y.I. did field and laboratory analysis. Y.M. did field observation. K.Y. organized meetings and gave advices.

Additional information

Competing financial interests: The authors declare no competing financial interests.
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